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(54) **Coextruded plastic film label for in-mold-labeling.**

(57) A label particularly adapted for use in in-mold labeling of blow-molded polyethylene containers comprising a heat activatable ethylene polymer adhesive layer and a surface printable layer with optional intermediate layers to provide interlayer adhesion and recycle of reground labels.

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COEXTRUDED PLASTIC FILM LABEL FOR IN-MOLD LABELING

The present invention relates to labels, especially to labels that are particularly adapted for use in in-mold labeling of blow-mold polyethylene containers. More particularly, the present invention relates to a coextruded plastic film label for use in such in-mold labeling applications.

The in-mold labeling of blow-mold polyethylene containers, particularly blow molded high density polyethylene containers allows the manufacturer to readily and inexpensively produce labeled containers directly from the molding operation without the need to apply adhesively backed labels in a subsequent step. By eliminating subsequent handling and adhesive applying operations capital investment and handling costs are substantially reduced. However, in-mold labeling equipment requires the cooperation of various mechanical devices which operate at relatively high speeds. Labels are supplied from a magazine and individually positioned by a mechanical or vacuum operated device

inside of a mold which is thereafter closed and the molded thermoplastic object prepared. Blow molding is a suitable molding process wherein a parison of heat plastified thermoplastic resin, most generally high density polyethylene, is extruded and then expanded to conform to the inner surface of a mold. The label, which also must conform to the inner surface of the mold, must be held in place until contacted with the heat plastified thermoplastic resin and thereafter must become attached to the resulting object by means of a heat activated adhesive contained on the surface of the label.

The entire procedure must be carried out in a continuous and rapid manner. The supply of labels from a suitable magazine, the positioning of the label within the mold, and the blow molding procedure must proceed in an uninterrupted manner in order to attain desired economical level of operation. Accordingly, labels for use in in-mold labeling processes are required to meet demanding performance requirements. Suitably, the labels must be sufficiently stiff that wrinkling or folding does not occur during handling by the automated equipment. Contrarywise, the labels are required to be sufficiently elastic or flexible that upon attachment to the desired blow molded object they will remain adhered thereto without splitting or separating despite bending, flexing, or squeezing of the finished container.

For certain applications the container, especially shampoo containers, laundry detergent containers, etc. may be exposed to high humidity or water. The adhesive used to adhere the label to such a container should desirably resist the effects of moisture

which may lead to delamination or wrinkling of the label.

A further requirement of in-mold labels is the presence of a suitable surface appearance. It is highly desirable from an esthetical and consumer acceptance point of view that the label not affect graphics or printing appearing on the surface thereof. Because thermoplastic scrap generated in the manufacture of coextruded films is desirably reused by recycling, i.e., regrinding and remelting, and incorporating the same into a separate inner layer of the film, it is highly desirable in the preparation of such labels that such scrap containing layer within the label not affect surface qualities of the label itself.

Finally, previously known in-mold labels prepared from cellulosic base stocks, such as paper, have proven undesirable in operation because the recycle of scrap blow-molded objects having such labels affixed thereto requires tedius mechanical means or solvents to first remove the labels. In order to reclaim or recycle such bottles, particularly defective bottles prepared during the blow-molding process, it is first necessary to remove the paper labels. Contamination by residual adhesive and small portions of unremoved paper may make it practically impossible to reclaim defective blow-molded containers bearing such labels. As an alternate means of disposal, defective containers must be burned or buried in landfills both of which practices are wasteful and undesirable.

It would be desirable to provide a polymeric label particularly adapted for use in in-mold labeling of blow-molded polyethylene containers.

In addition, it would be desirable to provide such a label for use in in-mold labeling which combines suitable properties of modulus of elasticity and flexibility, and at the same is adaptable to in-mold processing conditions and not degraded by flexing and handling of the subsequent container.

It would further be desirable to provide a label for use in in-mold labeling applications which is unaffected by moisture or high humidity environments.

Finally, it would be desirable to provide a label for in-mold labeling operations which does not have to be removed from such containers in order to recycle or regrind defective thermoplastic containers for subsequent incorporation into the thermoplastic.

According to the present invention there is now provided an adhesive label particularly adapted for use in the in-mold labeling of blow molded polyethylene containers comprising: 1) a heat activatable adhesive substrate layer of a polymer selected from: a) homopolymers of ethylene; b) copolymers of ethylene with one or more α-olefins having from 4 to 8 carbons; c) mixtures of a) and b); and d) mixtures of either a), b), or c) with up to 95 percent, preferably up to 50 percent, by weight of a copolymer of ethylene and from 1 percent to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxyl containing comonomer; 2) when the heat activatable adhesive substrate layer comprises a), b), or c), an interlayer

of a copolymer of ethylene and from 1 percent to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxyl containing comonomer; and 3) a surface printable layer comprising polystyrene.

The label desirably has a 1 percent secant modulus, measured according to American Society of Testing Methods (ASTM) test D-882, of from 150,000 to 600,000 lbs/in^2 (1.034 x 10^9 to 4.136 x 10^9 Pa) and a thickness of from 0.001 to 0.010 inch (.025 to 0.25 mm).

In one embodiment of the invention, the heat activatable adhesive substrate layer of the present invention desirably comprises a polyethylene polymer. Suitable polyethylene polymers include high density polyethylene, low density polyethylene, linear low density polyethylene and blends thereof. In this embodiment, it is desirable to insert an interlayer between the adhesive substrate layer and the surface printable layer in order to obtain suitable adhesion therebetween. In this case, the interlayer comprises a copolymer of ethylene and from 1 percent to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxy containing comonomer. Preferred copolymers contain from 5 percent to 25 percent by weight of the copolymerizable ethylenically unsaturated carboxyl containing comonomer.

As previously mentioned, the heat activatable adhesive substrate may also comprise the aforementioned polyethylene polymer blended with up to 95 percent, preferably up to 50 percent, by weight of a copolymer of ethylene and from 1 percent to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxyl

containing comonomer. In such an embodiment, the interlayer adhered between the adhesive substrate and the surface printable substrate may be eliminated. When amounts of carboxy containing comonomer greater than about 95 percent are employed, difficulty in film preparation and handling may be encountered.

Copolymerizable ethylenically unsaturated carboxyl containing comonomers for use herein include ethylenically unsaturated carboxylic acids and esters thereof having up to 12 carbons. Examples include acrylic acid, methacrylic acid, methylmethacrylate, butyl acrylate, vinyl acetate, and the like. A preferred ethylenically unsaturated carboxyl containing comonomer is vinyl acetate.

A preferred heat activatable adhesive substrate comprises a blend of from 50 to 75 percent of low density polyethylene and from 50 to 25 percent of a copolymer of ethylene and vinyl acetate, said copolymer containing from 5 to 25 percent vinyl acetate. A most preferred heat activatable adhesive substrate comprises from 60 to 70 percent low density polyethylene and from 40 to 30 percent ethylene/vinyl acetate copolymer.

The surface printable layer comprises polystyrene, and is adapted to receive printing ink, dyes, metal films, and the like and provide a smooth defect-free surface. Generally, pigment or fillers are provided in order to provide a suitable background for printing, and to hide refractive index changes in the other layers of the label or any blemishes or other defects appearing in the layer of reground and recycled polymer. A suitable surface printable layer comprises from 75 to 98 percent by weight of polystyrene and from

25 to 2 percent by weight of a suitable pigment, or filler. Examples of pigments include titanium dioxide, carbon black, and the like. Suitable fillers include calcium carbonate, silica, talc, and the like A particularly preferred pigment is titanium dioxide.

It is furthermore desirable in operation, that the labels of the present invention additionally include a layer interposed between the adhesive substrate and the surface printable layer that comprises reground and recycled thermoplastic material employed to prepare such labels. Significant waste occurs in the manufacturing of films used to prepare labels both in the generation of defective films, and in cuttings and trimmings left over from sizing and die-cutting of the desired shaped films and labels. Unless such waste is reground, remelted and included in the film stock, a significant economic loss is incurred. Accordingly, in a preferred embodiment of the present invention, a scrap or reground layer is interposed between the heat activatable adhesive substrate layer and the surface printable layer. Where the resulting structure comprises four layers, it is desirable that such reground layer be placed adjacent to the surface printable layer. The reground layer may comprise up to 50 percent by weight of reground label material with the balance comprising pigments, fillers and polystyrene. Because the reground layer may contain color bodies and have a marbled or swirled appearance, the surface printable layer desirably contains sufficient pigment so as to hide or cover over such defects appearing in additional layers of the label.

Because the labels of the present invention are adhered to the polyethylene blow-molded container by

melt adhesion of the heat activatable adhesive substrate an exceedingly strong adhesive bond is attained between the labels of the present invention and the blow-molded container. This bond is unaffected by the presence of water or high humidity which may cause delamination of adhesive applied in the form of dispersions and is also unaffected by flexing or bending of the underlying blow-molded container.

In addition, the absence of a noncompatible adhesive coating is believed to aid in producing a compatible blend upon regrinding and remelting of scrap containers. More particularly, defects such as swirls appearing in the reground layer are reduced by eliminating such an adhesive coating.

Thickness

In order to operate most smoothly in the in-mold labeling process, the label of the present invention preferably has a 1 percent secant modulus of from 200,000 to 400,000 pounds/in^2 (1.378×10^9 to 2.758×10^9 Pa) and a thickness of from 0.002 to 0.005 inches (0.05 to 0.127 mm).

Generally, any of the various layers in the label of the present invention may comprise from 10 to 50 percent of the total label thickness.

The label of the invention may be prepared by any suitable technique such as laminating of the various layers. Preferably a label stock is prepared by coextrusion of the various label layers utilizing coextrusion techniques previously known in the art. Suitably, the resins to be used in the various layers are separately heat plastified and extruded through a multi-feedblock die in the form of a thin sheet or

film. Alternatively, the extrusion is in the form of a tube which is later expanded by use of compressed air or an expansion mandrel to provide a thin film comprising the various layers according to the present invention. After cooling and slitting into a suitable width film, the labels are printed and die cut according to known techniques. Advantageously, because separately applied adhesive is not employed, the labels may be conveniently and economically prepared.

1. An adhesive label particularly adapted for use in the in-mold labeling of blow-molded polyethylene containers comprising: 1) a heat activatable adhesive substrate layer of a polymer selected from: a) homopolymers of ethylene; b) copolymers of ethylene with one or more α-olefins having from 4 to 8 carbons; c) mixtures of a) and b); and d) mixtures of either a), b), or c) with up to 95 percent, preferably up to 50 percent, by weight of a copolymer comprising ethylene and from 1 to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxyl containing comonomer; 2) when the heat activatable adhesive substrate layer comprises a), b), or c), an interlayer of a copolymer of ethylene and from 1 percent to 30 percent by weight of a copolymerizable ethylenically unsaturated carboxyl containing comonomer; and 3) a surface printable layer comprising polystyrene.

2. The label according to Claim 1, having a 1 percent secant modulus of from 150,000 to 600,000 lbs/in^2 (1.034×10^9 to 4.136×10^9 Pa) and a thickness of from 0.001 to 0.010 inch (.025 to 0.25 mm).

3. The label according to Claim 1, having a 1 percent secant modulus from 200,000 to 400,000 lb/in^2 (1.378×10^9 to 2.758×10^9 Pa).

4. The label according to Claim 1, having a thickness from 0.002 to 0.005 inches (0.05 to 0.127 mm).

5. The label according to Claim 1, wherein the heat activatable adhesive substrate comprises low density polyethylene.

6. The label according to Claim 1, wherein the carboxyl containing comonomer is vinyl acetate.

7. The label according to Claim 1, additionally containing a recycle layer of ground remelted label interposed between the heat activatable adhesive substrate and the surface printable layer.

8. The label according to Claim 1, wherein the surface printable layer additionally comprises from 30 to 2 percent by weight of a pigment or filler.